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(54) **TUBE REACTOR**

(75) Inventors: **Fred A. Conneway**, Titusville, FL (US);  
**Harvey E. Andresen**, Luling, LA (US);  
**Clarence P. Stadlwieser**, Sherwood Park  
(CA); **Donald L. Kurle**, St. Albans, WV  
(US); **Bernie B. Osborne**, Hurricane,  
WV (US)

(73) Assignee: **Dow Technology Investments LLC**,  
Midland, MI (US)

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**F28D 7/16** (2006.01)  
**C07D 301/10** (2006.01)

(52) **U.S. Cl.** ..... **422/201**; 549/534; 549/541

(58) **Field of Classification Search** ..... 422/201;  
549/534

See application file for complete search history.

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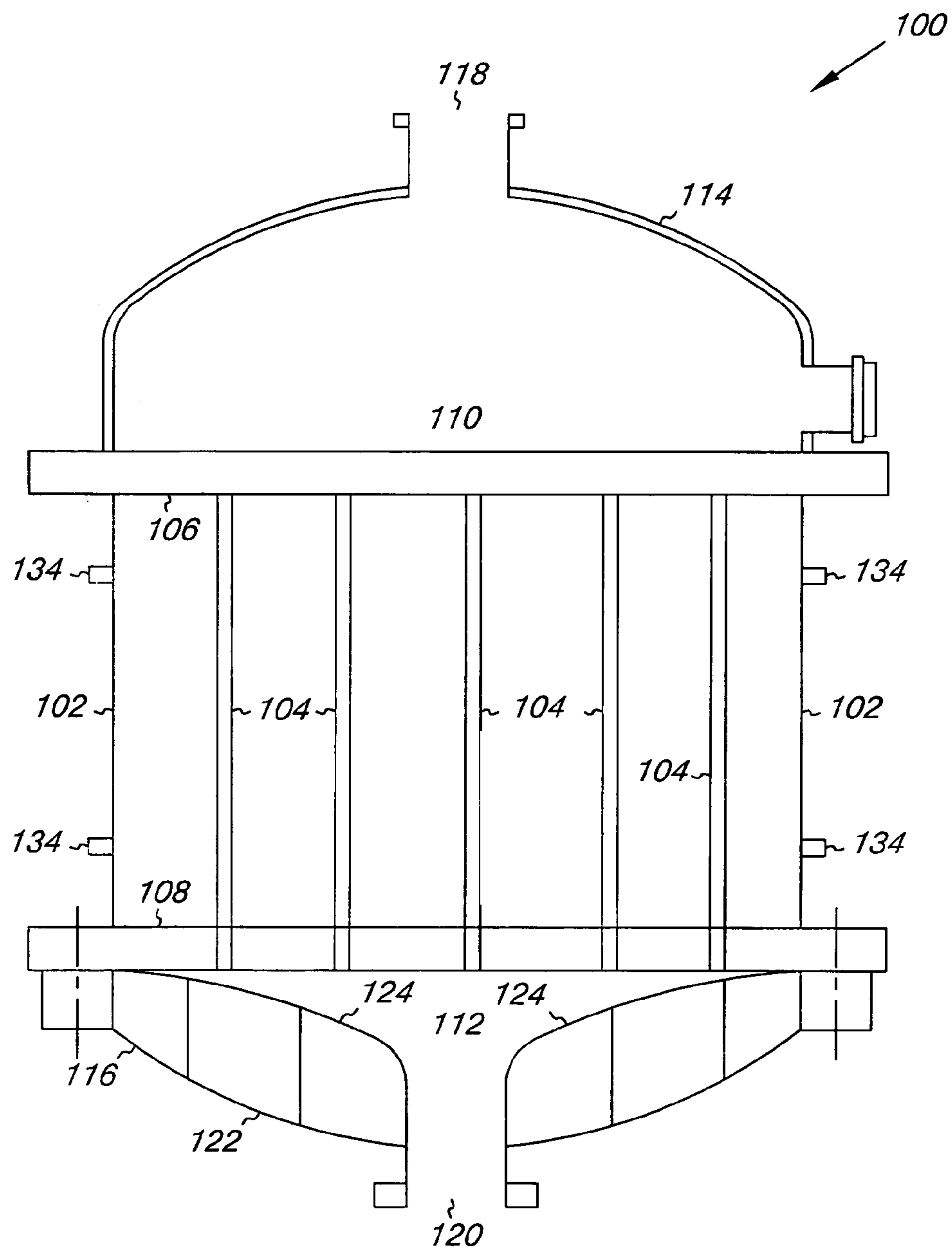
*Primary Examiner* — Bernard Dentz

(74) *Attorney, Agent, or Firm* — Brooks, Cameron &  
Huebsch, PLLC

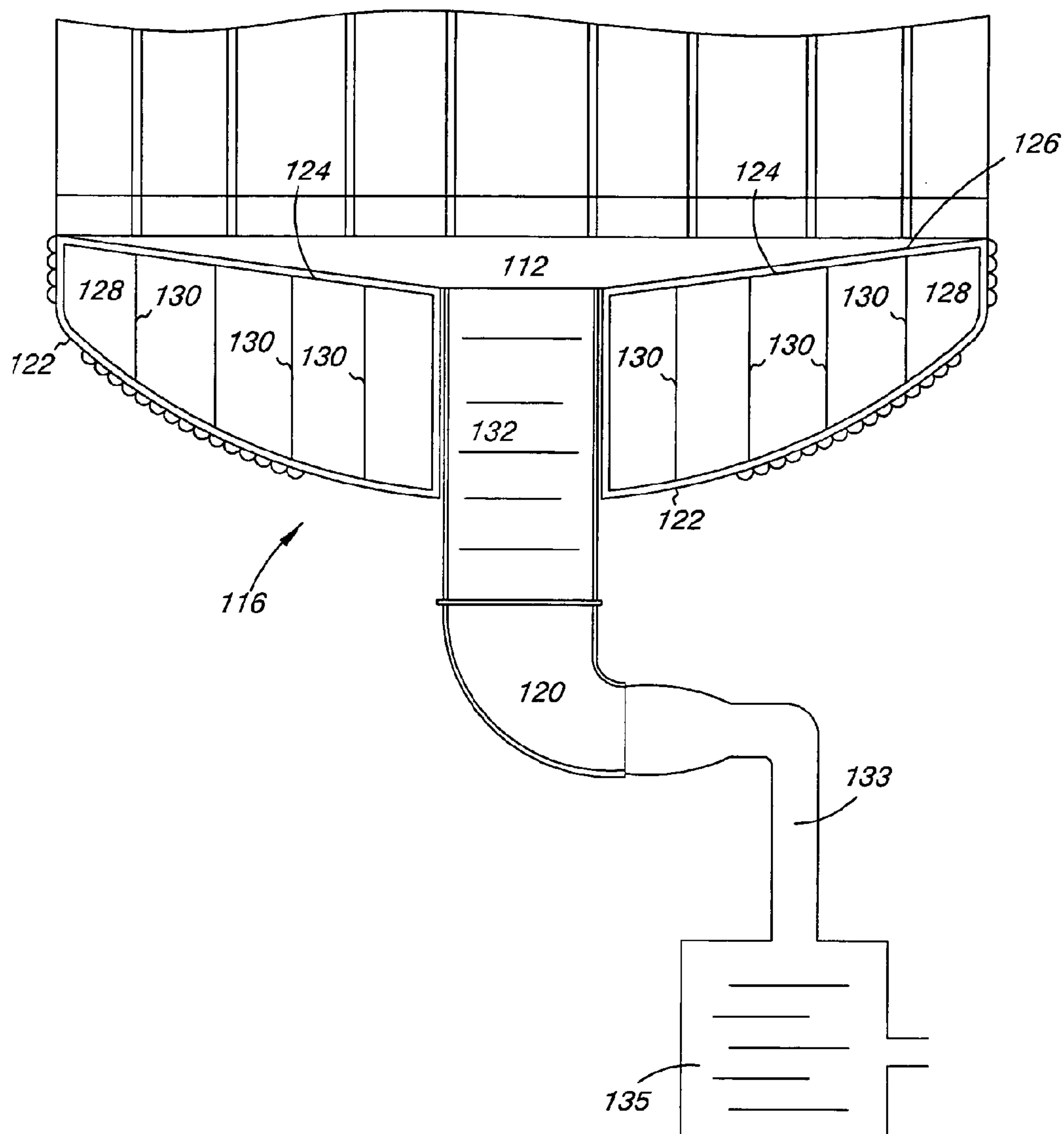
(57) **ABSTRACT**

Apparatus, methods, and processes are provided for a tube  
reactor including multiple, substantially parallel reaction  
tubes arranged within a tube reactor shell, the reaction tubes  
spaced apart such that a thermal fluid can flow between the  
tubes and transfer heat between the tubes and the thermal fluid  
during operation; an inlet head defining an inlet head space,  
where the inlet head space is in fluid communication with an  
inlet end of the reaction tubes, and an outlet head including an  
outlet head shell and at least one insert positioned within an  
outlet head space defined by the outlet head, where the insert  
defines a reduced volume outlet head space relative to that  
defined by the outlet head, and where the reduced volume  
outlet head space is in fluid communication with an outlet end  
of the reaction tubes and in fluid communication with a reac-  
tor outlet.

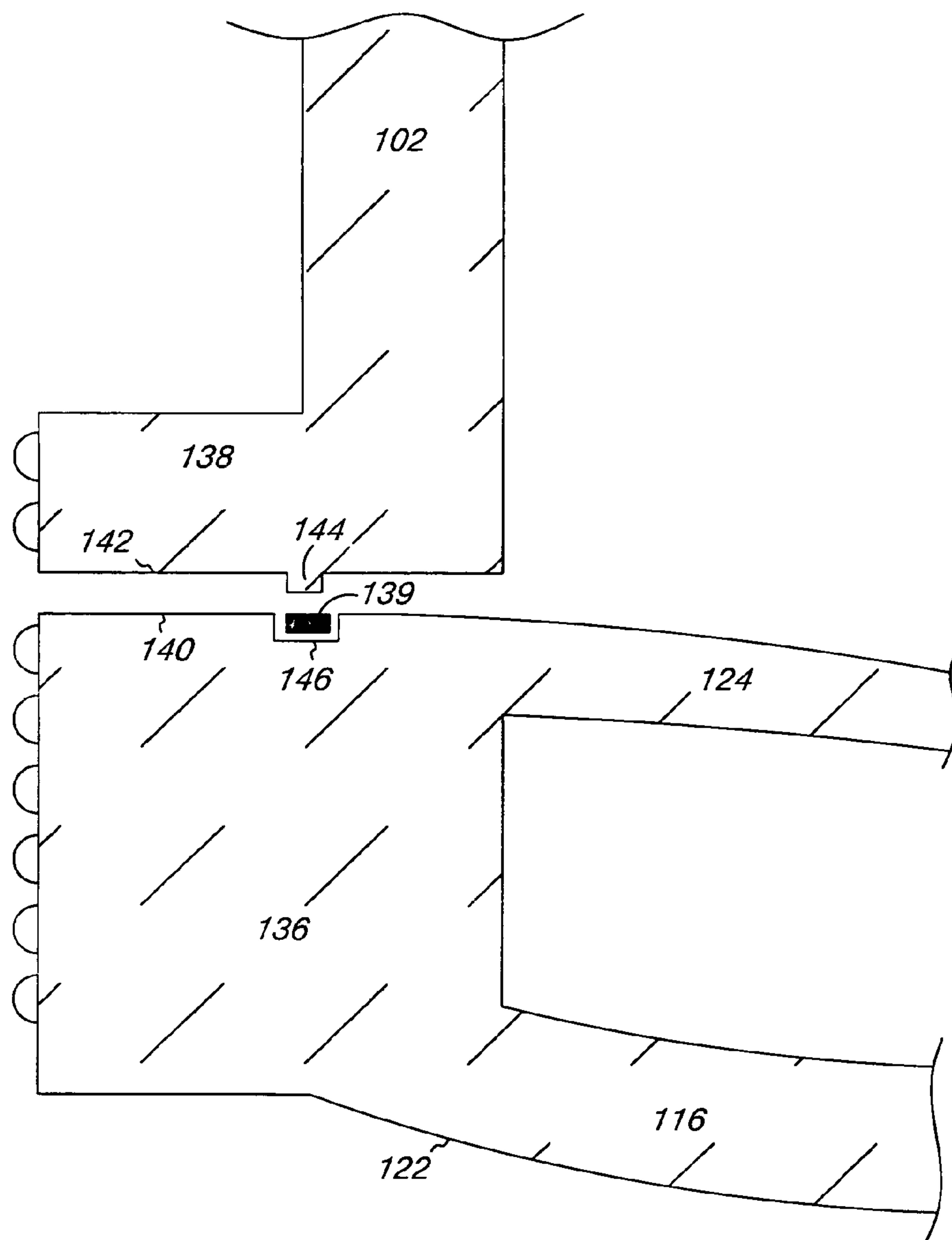
**18 Claims, 7 Drawing Sheets**



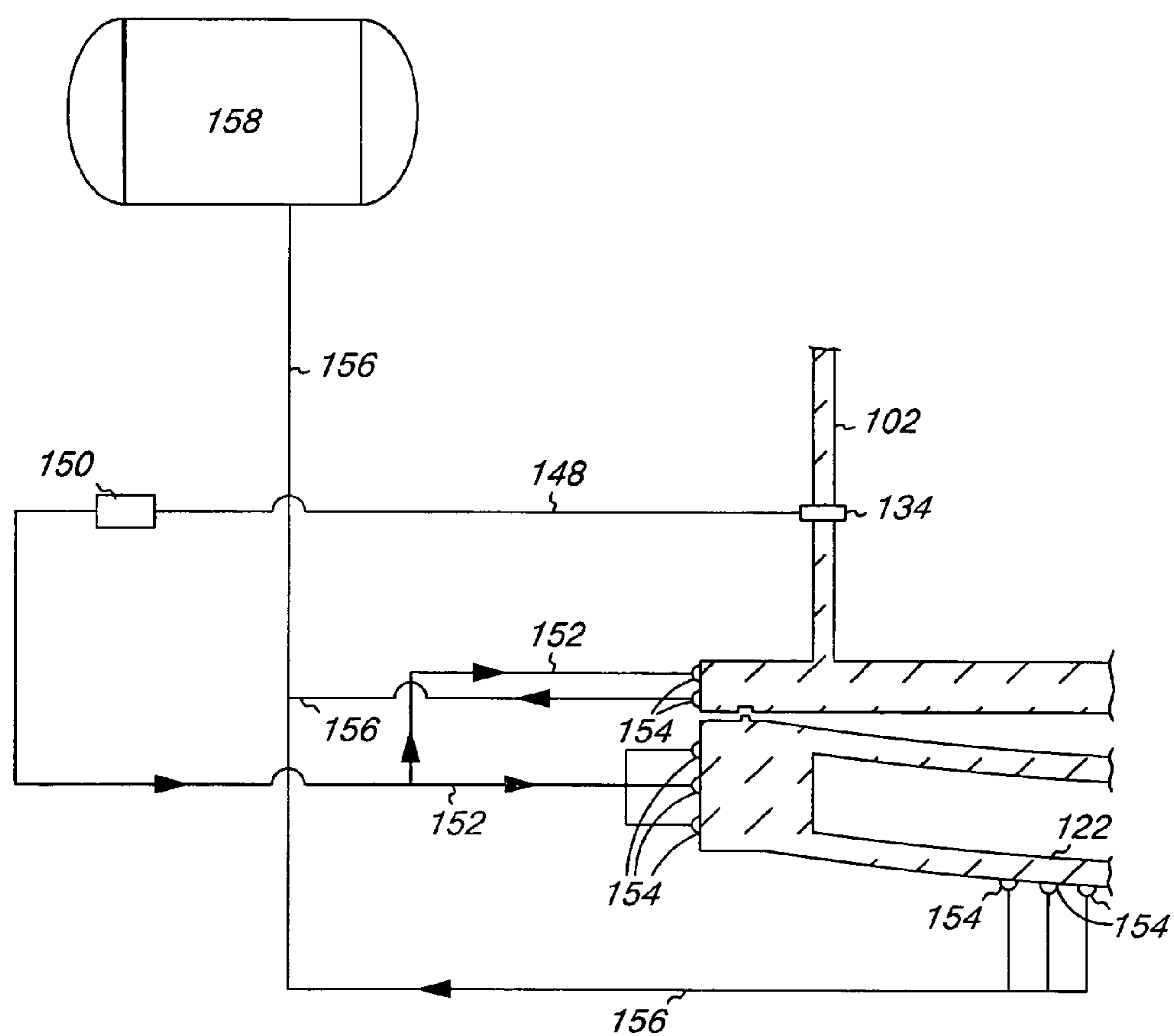
*Fig. 1A*



*Fig. 1B*



*Fig. 1C*



*Fig. 1D*

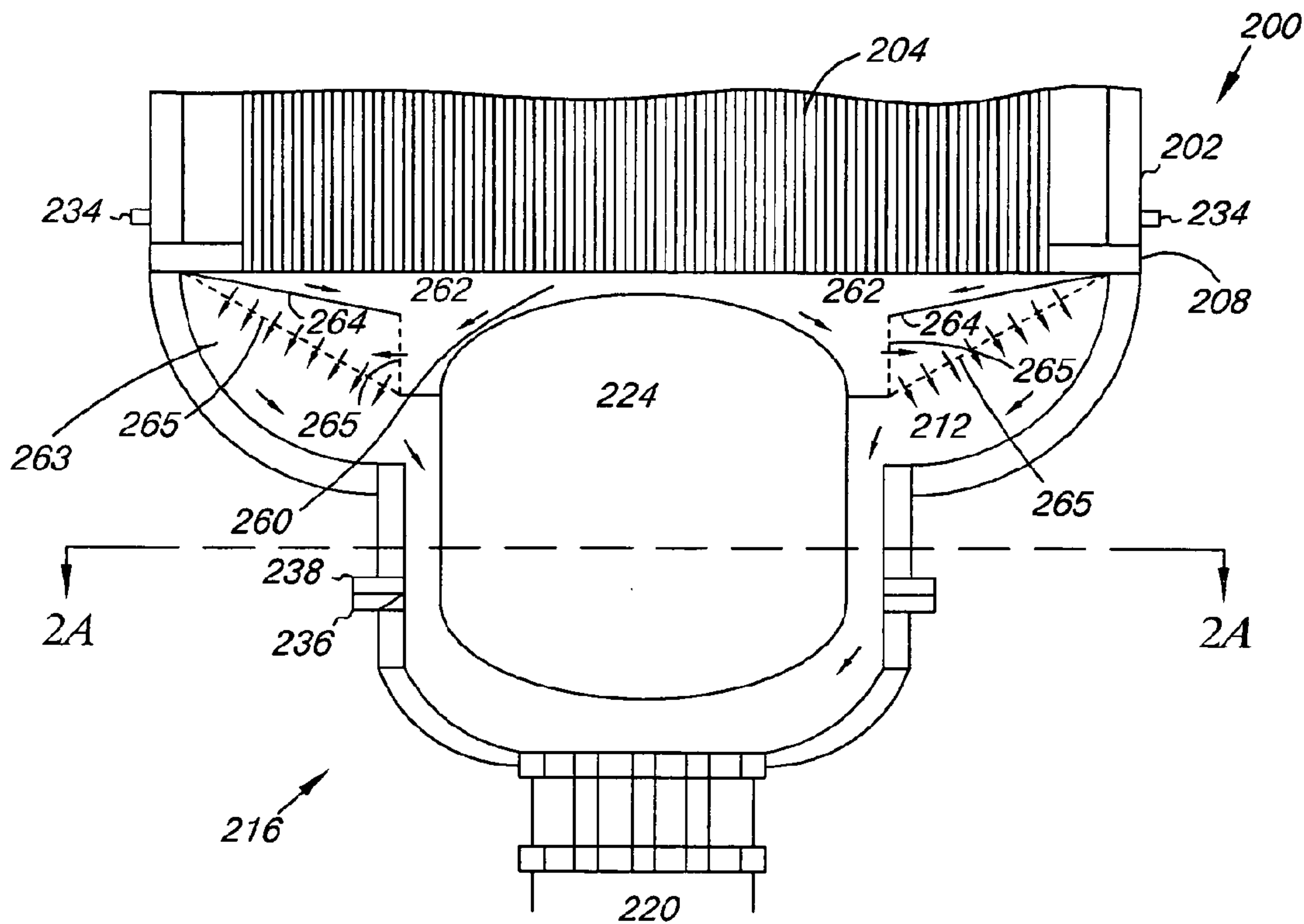


Fig. 2

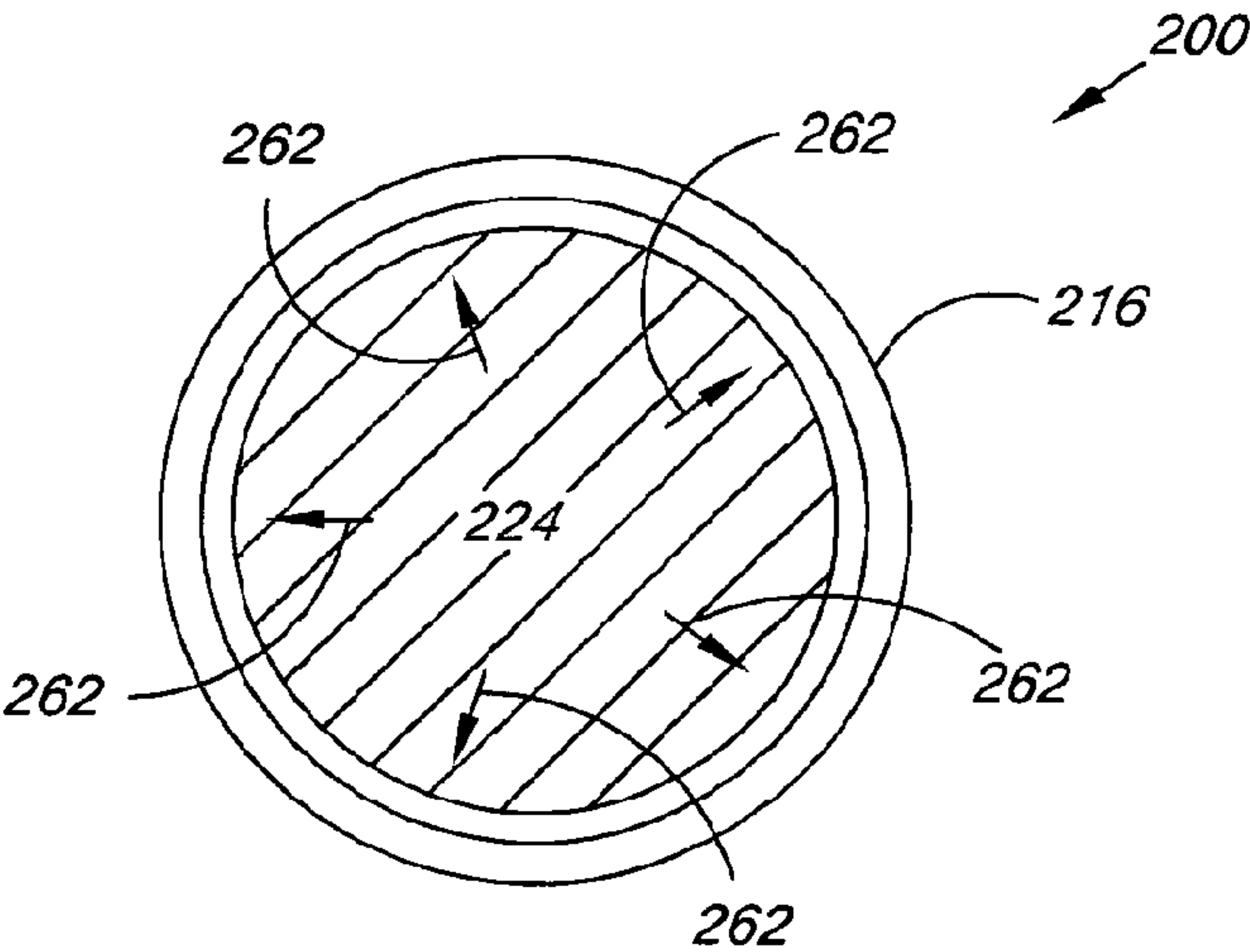
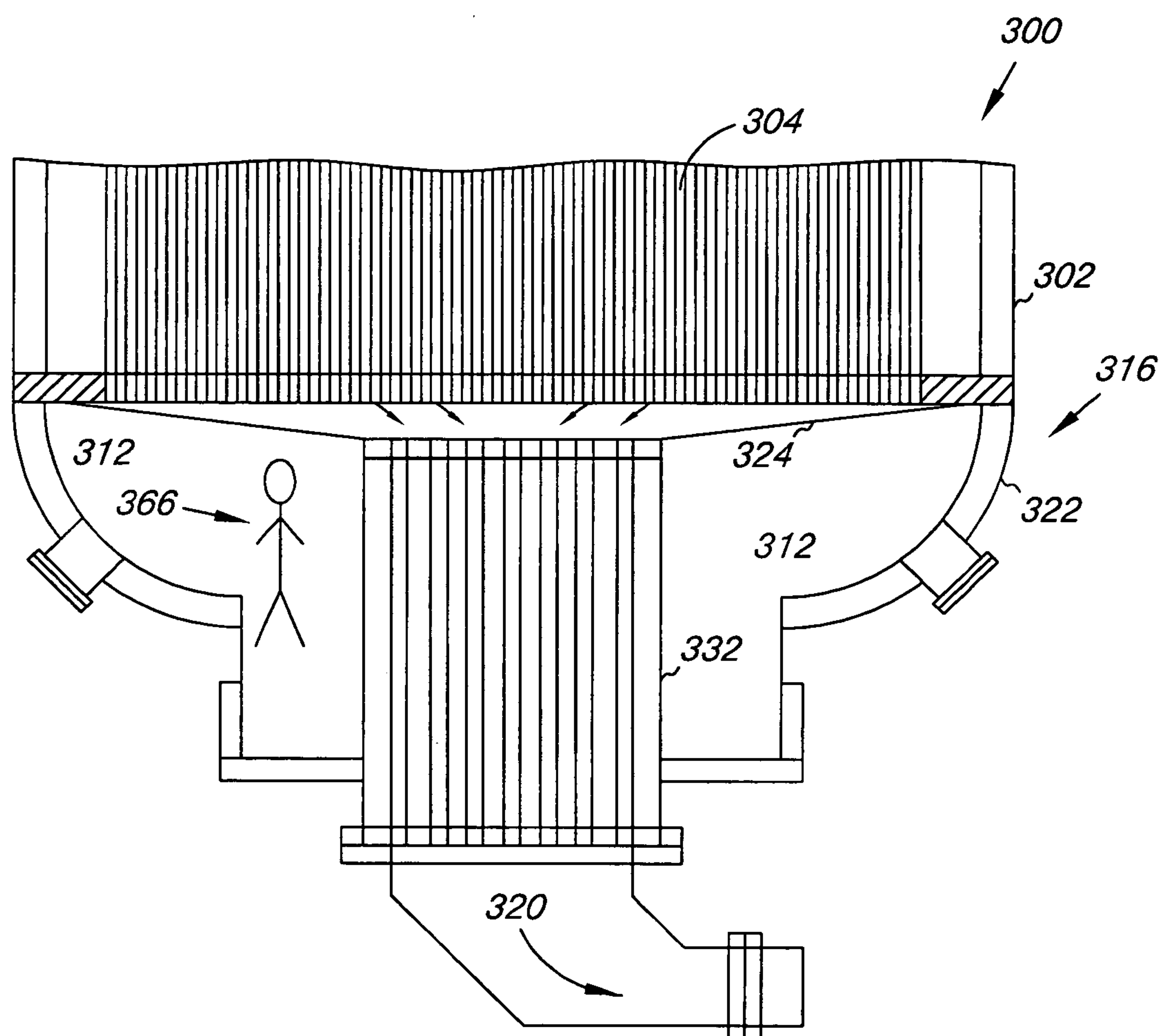
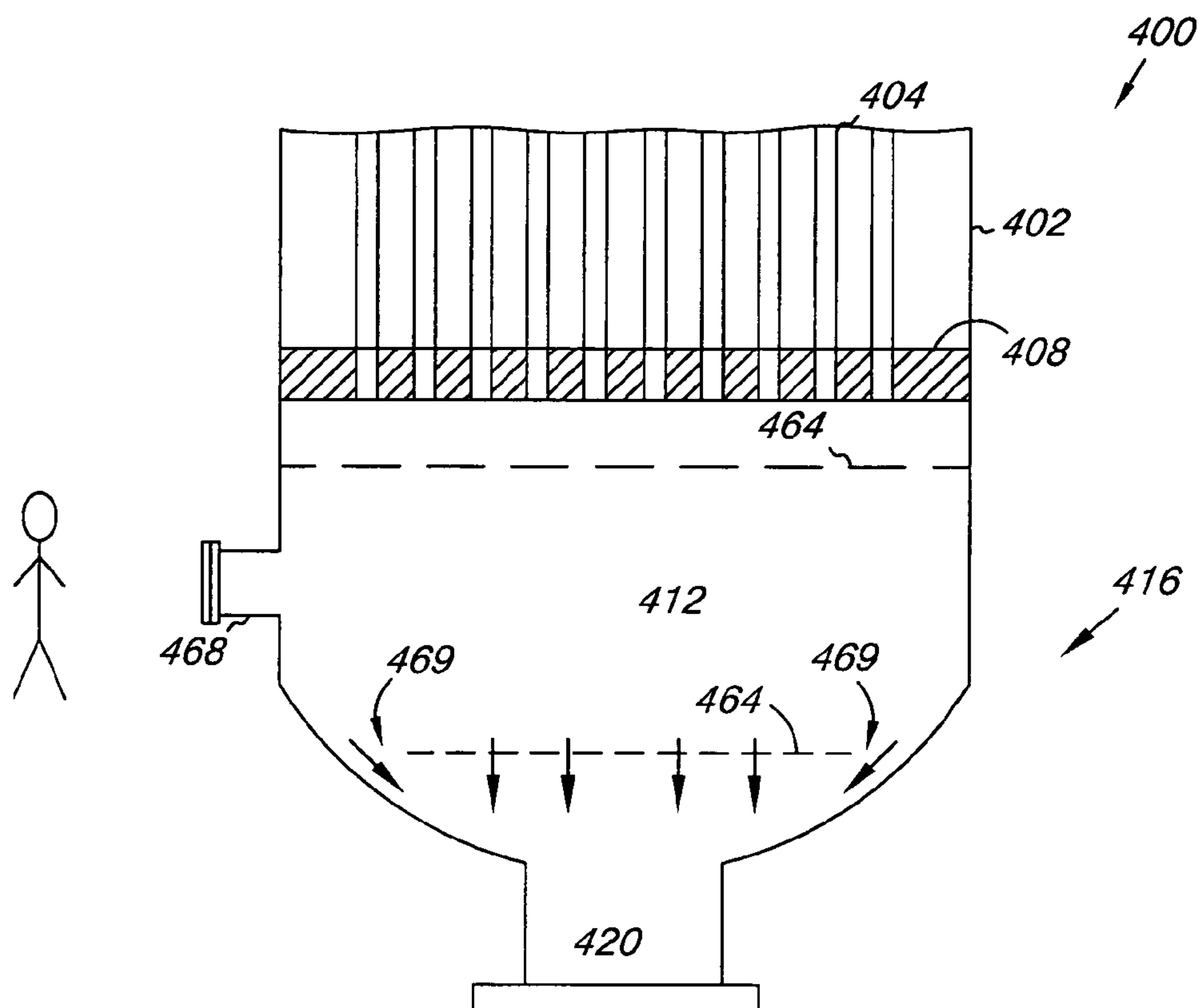


Fig. 2A





*Fig. 3*



*Fig. 4*



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## TUBE REACTOR

This application is a National Stage application under 35 U.S.C. 371 of PCT/US2008/013501, and published as WO 2009/116977 on Sep. 24, 2009, which claims priority in part from U.S. Provisional Application Ser. No. 61/008,062 filed Dec. 18, 2007, the entire content of which is incorporated herein by reference.

## BACKGROUND

The oxidation of ethylene to ethylene oxide can be accomplished by reaction with air or oxygen in the presence of a silver catalyst mounted on a suitable carrier, with the optional addition of small amounts of certain chlorinated hydrocarbons which serve to moderate the catalyst. Under normal operating conditions, the oxidation reaction produces mostly ethylene oxide, with carbon dioxide and water as side products. The reaction is exothermic.

The construction of the reactor where the oxidation is carried out can be similar to a vertical shell and tube heat exchanger. The tubes are filled with the catalyst while the shell side of the reactor is provided with a coolant. Suitable pumps, heat exchangers, separator drums, and/or condensers can be employed to condense, or separate, and recirculate the coolant.

In some cases, without careful temperature control, the oxidation reaction can proceed to the formation of carbon dioxide and water vapor. This not only reduces the yield, but is a much more exothermic reaction which can cause "hot spots" within the reactor and a consequent increase in the demands made upon the cooling system. In addition, the "hot spots" can increase local temperatures and can spread rapidly to the reactor outlet, leading to a bulk gas phase ignition, referred to herein as a "decomp".

## SUMMARY

The present disclosure relates to tube reactors for conducting exothermic reactions such as alkene oxidation reactions. Embodiments are adaptable to commercial scale ethylene oxide production.

As used herein, a "hot spot," is an uncontrolled, gas-phase reaction occurring inside of a catalyst bed located in a reactor tube in which oxygen is depleted.

Embodiments of the present disclosure include a tube reactor including multiple, substantially parallel reaction tubes arranged within a tube reactor shell, the reaction tubes being spaced apart such that a thermal fluid can flow between the tubes and transfer heat between the tubes and the thermal fluid during operation. The tube reactor also includes an inlet head positioned adjacent the tube reactor shell defining an inlet head space, where the inlet head space is in fluid communication with an inlet end of the reaction tubes and an outlet head positioned adjacent to the tube reactor shell. The outlet head includes an outlet head shell and at least one insert positioned within an outer head space defined by the outlet head, where the insert defines a reduced volume outlet head space relative to that defined by the outlet head, and where the reduced volume outlet head space is in fluid communication with an outlet end of the reaction tubes and in fluid communication with a reactor outlet.

Gas residence times in the outlet head can be reduced when embodiments of the tube reactor of the present disclosure are operated. The reduced gas residence times permit hot reaction gases to be passed more quickly to a cooling step and can help to reduce the formation of reaction by-products. Areas of

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stagnant gases are also reduced or eliminated, again helping to reduce by-product formation and, in cases in which the process gases are flammable, helping to reduce the risks of fire due to the development of "decomps" in the outlet head space.

## BRIEF DESCRIPTION OF THE DRAWINGS

FIG. 1A is a front view, in section, of an embodiment of a tube reactor of the present disclosure.

FIG. 1B is a front view, partially in section, of an outlet head with insert for use with embodiments of the present disclosure.

FIG. 1C is a front sectional view of a flange region of a tube reactor shell and removable outlet head in accordance with embodiments of the present disclosure.

FIG. 1D is a schematic view of a system for circulating thermal fluid from the tube reactor shell to a flange region of the tube reactor shell and removable outlet head in accordance with embodiments of the present disclosure.

FIG. 2 is a front view, partially in section, of an embodiment of the present disclosure.

FIG. 2A is a top sectional view, taken along lines 2A-2A of FIG. 2, showing an outlet head with insert in accordance with embodiments of the present disclosure.

FIG. 3 is a front view, in section, of an outlet head according to embodiments of the present disclosure.

FIG. 4 is a front view, in section, of an outlet head according to embodiments of the present disclosure.

## DETAILED DESCRIPTION

Embodiments of the present disclosure include apparatuses, methods, and processes for tube reactors and the operation of tube reactors. For purposes of illustration, the present disclosure is discussed in its application to the production of ethylene oxide, however, it is understood that embodiments of the present disclosure are not limited to this application.

FIGS. 1A-1D illustrates an embodiment of a tube reactor **100** according to the present disclosure. The tube reactor **100** includes an exterior shell **102** that encloses a space that contains reactor tubes **104**. In some embodiments, the reactor tubes **104** can be arranged substantially parallel to each other and can be oriented vertically with the reactor tube outlets at the bottom of the tube reactor **100**. In some embodiments, the tube reactor **100** can be oriented with the tube outlets at the top. Also, the tube reactor **100** can be oriented horizontally, or at some angle to the horizontal, if desired for specific applications.

The reactor tubes **104** are spaced apart from each other in order to permit thermal fluid to circulate between adjacent reactor tubes **104** to transfer heat between the thermal fluid and the reactor tubes **104**. As illustrated in FIG. 1A, the tube reactor **100** contains a small number of reactor tubes for purposes of illustration. Also, the spacing between the tubes **104** is exaggerated, again for purposes of illustration. In various embodiments, the tube reactor **100** can contain reactor tubes **104** in a range of hundreds to thousands.

In some embodiments, the reactor tubes **104** can be held in place within the tube reactor shell **102** via a top tubesheet **106** and a bottom tubesheet **108**. The tubesheets **106**, **108** have openings to allow process gases to enter the reactor tubes **104** from the inlet head space **110**. Alternatively, the reactor tubes **104** can extend through the top tubesheet **106** into the inlet head space **110**. Similarly, the bottom tubesheet **108** has openings to permit process gases to enter an outlet head space



112 from the reactor tubes 104. Again, the reactor tubes 104 can extend through the bottom tubesheet 108 into the outlet head space 112.

In some embodiments, various types of additional supports can be provided between the top tubesheet 106 and the bottom tubesheet 108. For example, additional supports can include additional tubesheets, various types of brackets, baffles, and the like.

As shown in FIG. 1A, the tube reactor 100 also includes an inlet head 114 and an outlet head 116 (e.g., in embodiments where the reactants have a downward flow during operation). As shown, the inlet head 114 is positioned adjacent the tube reactor shell 102 at a top position and the outlet head 116 is positioned adjacent the tube reactor shell 102 at a bottom position. Embodiments can also include the inlet head 114 positioned at the bottom position and the outlet head 116 positioned at the top position.

The inlet head 114 defines the inlet head space 110. Reactants can be introduced into the tube reactor 100 through an inlet conduit 118, where the reactants enter the inlet head space 110. As illustrated, from the inlet head space 110, the reactants can enter the tubes 104 and flow downwardly through the tubes 104, where the reactants come into contact with a catalyst and the desired reaction occurs. Reaction product gases thus exit the bottom of the tubes 104 and enter the outlet head space 112, where the reaction product gases from the various tubes 104 mix and enter the outlet conduit 120.

Commercial scale tube reactors of the type illustrated in FIG. 1 can be very large, for example, containing hundreds or thousands of tubes and having a shell diameter in a range of three (3) meters to as large as twenty (20) meters. Because of the large size, the inlet head 114 and outlet head 116 are designed with a rounded cross-section, as illustrated in FIG. 1A. The curved shape of the inlet head 114 and the outlet head 116 can permit the heads to have the needed structural strength at reasonable masses. However, because of the architecture of the heads 114, 116, the volume of the inlet head space 110 and the outlet head space 112 can be significant. Large head spaces result in larger residence times within the head spaces. In addition, circulation of reaction products within the head spaces is not uniform and as a result, hot spots can form in regions where pockets of stagnant and/or recirculating gas exist. In the hot spot regions, unwanted side reactions can occur as well as an increased risk of decomps.

To reduce the occurrence of the formation of hot spot regions, embodiments of the present disclosure seek to reduce the volume of the outlet head space 112, provide for initial rapid mixing of the reaction product gases exiting the reactor tubes 104, and/or provide for the redistribution of the reaction gases in the outlet head space 112. As such, in some embodiments, the residence time of the reaction product gases in the outlet head 112 can be minimized. In addition, the residence time distribution, or the probability distribution function that describes the amount of time the reaction product gases can spend inside the outlet head space 112, can be tightened to reduce a long residence time "tail."

In various embodiments, local hot spots formed as reaction product gases exit some of the reactor tubes 104 can be blended rapidly with cool reaction product gases exiting other reactor tubes 104, preventing ignition and/or unwanted side reactions.

Because of the size of the tube reactors 100, it is not practical to reduce the outlet head space volume simply by using a smaller outlet head. The mass of such an outlet head would be much greater than that of the curved outlet head 116 as shown in FIG. 1, thus the cost and weight associated with

such a head makes it impractical. As such, as illustrated in FIG. 1A, the outlet head 116 includes an outlet head shell 122 and an insert 124. Together with the bottom tubesheet 108, the insert 124 can define the volume of the outlet head space 112.

As shown, the volume of the outlet head space 112 is considerably smaller than the volume that, in the absence of the insert 124, would be defined by the bottom tubesheet 108 and the outlet head shell 122. The smaller volume can reduce stagnant areas and can increase flow velocities through the outlet head space 112, while also decreasing residence times of the reaction product gases in the outlet head space 112. For example, the process gases can flow through the bottom tubesheet 108 at a velocity of approximately twenty-five (25) feet per second or more and the residence time of the reaction product gases can be approximately 0.1 second or less. Moreover, the flow pattern of the reaction product gases contain no large scale eddies.

In some embodiments, the insert 124 can reduce the outlet head 116 volume by at least twenty-five (25) percent as compared to an outlet head 116 of the same configuration without the insert 124. In various embodiments, the insert 124 reduces outlet head 116 volume by at least fifty (50) percent. The insert 124 may reduce outlet head 116 volume by up to sixty (60) percent, by up to seventy-five (75) percent, by up to eighty-five (85) percent, or by up to ninety (90) percent or more.

In addition, flow rates (i.e., gas velocities) through the outlet head 116 can increase with greater reductions in outlet head 116 volume. Also, residence times in the outlet head 116 decrease correspondingly.

In some cases, too great a reduction in outlet head 116 volume can make it difficult to achieve high bulk flow rates at reasonable operating pressures since the path through the outlet head 116 can become restricted. Thus, the upper limit on how much the outlet head 116 volume can be reduced may, in some cases, be limited by desired bulk flow rates and/or operating pressures.

FIG. 1B illustrates an embodiment of the outlet head 116 according to the present disclosure in greater detail. The insert 124 is shown sloping downwardly from an edge region 126 towards the outlet 120. As such, the height of the outlet head space 112 is greater near the center than at the edges of the insert 124. This configuration can improve flow rates (e.g., both gas velocities and bulk flow rates) and can help eliminate stagnant areas, particularly near the outer edges of the outlet head space 112.

The outlet head shell 122 also can curve downwardly from the edges toward the outlet 120, but can curve at a more gradual rate as compared to the insert 124. The difference in curvature can cause the outlet head shell 122 to be displaced away from the insert 124, forming a space 128 between the outlet head shell 122 and the insert 124. In such embodiments, reaction product gases do not come into contact with the outlet head shell 122, except perhaps at the region adjacent to the outlet 120, and the outlet head shell 122 is somewhat thermally isolated from the process fluids.

In some embodiments, the outlet head shell 122 is designed to bear approximately all the weight of the outlet head 116 as well as other applied stresses. In such embodiments, the external curvature of the outlet head shell 122 can permit the outlet head shell 122 to be made using reasonable masses of construction materials. In addition, the insert 124 bears little weight or stress, and thus can be made from relatively thin materials in order to conserve mass.

In some embodiments, the insert 124 can be self-supporting. In addition, in various embodiments the insert 124 can have a light weight construction obtained by using a thinner



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material for the insert **124** as compared to a self-supporting insert **124**. In such embodiments, the light weight insert **124** can include additional supports, such as a number of support cylinders **130**, which can traverse the space **128** and transfer the load from the insert **124** to the outlet head shell **122**. The insert **124** can be formed of a metal, polymer, and/or a ceramic material, provided that the material of construction is capable of withstanding process temperatures, does not catalyze reactions of the process gases and/or otherwise react with the process gases, and is otherwise chemically stable under the conditions of the particular process.

As illustrated in FIG. 1B, in some embodiments, the outlet head **116** can cool reaction product gases within the outlet head **116**. In various embodiments, a method of cooling the process gases can use a thermal fluid to absorb heat from the reaction product gases. Also, in some embodiments, as shown, reaction product gases can be cooled within the outlet head space **112** by passing the process gases through a heat exchanger **132** before the reaction product gases exit the reactor tube **100** via the outlet **120**.

In some embodiments, the heat exchanger **132** can capture the heat from the process gases and reuse the heat elsewhere. In some cases, the partially heated thermal fluid can be circulated to the tube reactor **100** to provide cooling to the reactor tubes. In some embodiments, the partially heated thermal fluid can be circulated to a flange system of the tube reactor shell **102** and outlet head **116** to equilibrate the temperatures of each, as discussed further herein.

In some embodiments, the ability to cool the reaction product gases immediately downstream of the point at which they exit the reactor tubes can be beneficial in reducing the formation of unwanted reaction byproducts. This is particularly true in alkene oxidation processes (e.g., ethylene oxide processes), in which aldehydes and other unwanted byproducts often form in the hot reaction product gases that exit the reaction tubes before the gases can be cooled. In the embodiment shown in FIG. 1B, the presence of the heat exchanger **132** within the outlet head **116** can permit the reaction product gases to be cooled rapidly, and can reduce byproduct formation as compared to an outlet head **116** without the heat exchanger **132**, particularly in ethylene oxide production processes.

In some embodiments, as shown in FIG. 1B, the outlet **120** located downstream of the heat exchanger **132** can be further connected by, for example, piping **133**, to a second heat exchanger **135**. The reaction product gases can pass through the heat exchanger **132**, through the outlet **120**, and through the pipe **133** to the second heat exchanger **135**. The second heat exchanger **135** can further cool the reaction product gas coming from the outlet **120**.

In some embodiments, the second heat exchanger **135** is close-coupled to the heat exchanger **132**. As used herein, "close-coupled" refers to coupling the second heat exchanger **135** at as short of a distance as possible from the heat exchanger **132**. In some embodiments, the length of the piping **133** can be less than one hundred (100) feet, more preferably, less than fifty (50) feet, and still more preferably, less than ten (10) feet. In addition, the diameter of the piping **133** can be as small as possible, preferably less than one hundred (100) inches in diameter and more preferably less than sixty (60) inches in diameter. Minimizing the distance between the heat exchanger **132** and the second heat exchanger **135** can reduce the residence time of the gas in the zone between the two heat exchangers and can further reduce the formation of reaction by-products.

In some embodiments, the reaction product gases can be cooled entirely within the outlet head **116**. In various embodi-

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ments, the process gases can be cooled partially within the outlet head **116**. For example, the heat exchanger **132** can be positioned entirely or only partially within the outlet head **116**. In embodiments where the heat exchanger **132** is positioned partially within the outlet head **116**, the heat exchanger **132** can extend outwardly beyond the confines of the outlet head **116** in order to provide a larger region for heat transfer, more thoroughly completing the cooling.

As shown in FIG. 1A, in operation, process gases enter the inlet head space **110** through the inlet **118**. The process gases become distributed among and enter the reactor tubes **104**. The reactor tubes **104** are, in some embodiments, at least partially filled with a catalyst for the desired reaction of the process gases. As the process gases flow through the reactor tubes **104**, they can react in the presence of the catalyst. The reaction product gases then flow out of the reactor tubes **104** and into the outlet head space **112**. In the embodiment shown in FIG. 1A, the reaction product gases exit the outlet head space **112** through the outlet **120**.

In some embodiments, the reaction occurring within the reaction tubes **104** can be highly exothermic, which is the case, for example, for an ethylene oxide oxidation reaction. In various embodiments, the exotherm can be controlled by circulating a thermal fluid within the shell **102** and around the exterior of the tubes **104**. In some embodiments, water is the cooling fluid. Water can be boiled to produce steam, therefore additional cooling can be provided since additional heat required to vaporize the steam is removed from the system.

Thermal fluid can be introduced through one or more ports **134**, through which it enters the tube reactor **100** and flows through the spaces between the reactor tubes **104**, coming into contact with the reactor tubes **104**, and providing either heating or cooling to the reaction process.

In some embodiments, the thermal fluid can provide cooling, for example, when the reaction occurring at the catalyst within the reactor tubes **104** is exothermic. However, in other embodiments, the thermal fluid can heat the reactor tubes **104** to a temperature that is needed to produce a desired reaction at a desired reaction rate. In various embodiments, when the reaction is exothermic, the thermal fluid can provide heat, for example, during start-up to initiate the reaction or to provide process control until steady-state conditions are achieved. Thermal fluid can be withdrawn from the tube reactor **100** through one or more ports **134**.

In some embodiments, the outlet head **116** is removably affixed to the tube reactor shell **102** in order to permit access to the interior of the tube reactor **100** for purposes such as maintenance, repair, and/or catalyst removal and/or replacement. In some embodiments, the outlet head **116** and tube reactor shell **102** include a flange system, as shown in FIG. 1B and in an enlarged view in FIG. 1C.

As shown in FIG. 1C, a suitable flange system includes mating circumferential flanges **136** and **138**, where the mating circumferential flanges **136**, **138** are attached to the bottom portions of the outlet head **116** and the tube reactor shell **102**, respectively. The flanges **136**, **138** can have mating surfaces **140**, **142** where the flanges **136**, **138** meet when joined. In some embodiments, the flanges **136**, **138** can be an extension of the tube reactor shell **102** and/or tubesheet **106**.

Optionally, the mating surfaces **140**, **142** can include one or more additional mating features, which can assist in aligning the outlet head **116** and the tube reactor shell **102** in the correct relative position during assembly. One such type of alignment feature is a circumferential tongue-and-groove assembly. As shown in FIG. 1C, the flange **138** can include a raised tongue **144** which can fit into a corresponding recessed groove **146** in the flange **136**. In some embodiments, the



groove **146** can be somewhat wider than the tongue **144** for easier assembly. In various embodiments, the raised tongue **144** can be located in flange **138** and the corresponding recessed groove **146** can be located in flange **136**. In addition, although not shown in FIG. 1C, a gasketing material **139** can be included within the groove **146** to aid in sealing the tube reactor **100**. Also, other types of guides can be incorporated into the flange system, including guide pin-and-hole arrangements.

As discussed herein, the diameter of the tube reactor **100** can range, for example, from two (2) meters to twenty (20) meters or more. Due to the size of the reactor **100**, the relative thermal expansion or contraction of the tube reactor shell **102** and the outlet head **116** can become an important design consideration for a number of reasons. For example, the outlet head shell **122** of the outlet head **116** can be at least partially thermally isolated from the reaction product gases due to the presence of the insert **124**. Therefore, the outlet head shell **122** can be, periodically, at a significantly different temperature than the tube reactor shell **102**, especially during start-ups, shut-downs, and during process upsets. The difference in temperature can result in different degrees of thermal expansion of tube reactor shell **102** and outlet head shell **122** and can create significant stresses due to the large diameter of the equipment. The stresses can cause distortion and break the seal at the flange. In some cases, the equipment can be damaged by the thermal stresses.

Due to the possibility of thermal stresses causing damage to the equipment, embodiments of the present disclosure maintain the outlet head shell **122** at a temperature approximately equal to the temperature of the tube reactor shell **102**. The temperatures can be maintained approximately equal using a number of methods. For example, heating and/or cooling can be provided by applying forced heated or cooled gas to the surface of the outlet head shell **122**. Heating can also be provided using electrically resistive coils included in the outlet head shell **122**.

In some embodiments, thermal fluid can be introduced to the outlet head shell **122**. The thermal fluid can be applied via a jacket (e.g., located on the interior and/or exterior surface of the outlet head shell **122**), and/or through conduits that pass within the outlet head shell **122**, and/or on the outlet head shell's **122** interior and/or exterior surface.

In various embodiments, a temperature control system can be included, for example, including temperature measuring devices which operate the temperature control system in response to the temperature of the outlet head shell **122**. Also, a variety of pumping or actuation systems can be included which can produce a flow of the thermal fluid in response to, for example, temperature differences between the outlet head shell **122** and the tube reactor shell **102**.

In some embodiments, the method of maintaining the temperature of the outlet head shell **122** and the tube reactor shell **102** at approximately equal temperatures includes contacting the outlet head shell **122** with a thermal fluid that is also used to control the temperature inside the tube reactor **100**. As discussed herein, a thermal fluid can be introduced into the tube reactor **100** to provide temperature control. For example, the thermal fluid can be introduced to control heat generated in exothermic reactions that take place within the reactor tubes **104** and/or be used to supply heat to a reaction.

During operation, the temperature of the tube reactor shell **102** can closely approximate the temperature of the thermal fluid inside the tube reactor **100** since the tube reactor shell **102** is in contact with the thermal fluid. Therefore, by circulating the thermal fluid into contact with the outlet head shell **122**, the temperature of the outlet head shell **122** can be

brought to approximately the same temperature as the tube reactor shell **102**. Thermal expansion and/or contraction of the two segments can therefore be closely matched, and stresses, distortion, and/or leakages associated with differential thermal expansion between the two sections can be minimized.

In addition, in order to match thermal expansion and contraction of the tube reactor shell **102** and the outlet head shell **122**, the two segments can be formed of the same material or formed of different materials that have similar coefficients of linear thermal expansion.

Contacting the thermal fluid from the reaction tube **100** with the outlet head shell **122** decreases the need for a separate temperature control system since a close temperature match between the tube reactor shell **102** and the outlet head shell **122** can be obtained simply due to the circulation of the same thermal fluid to both components.

FIG. 1D illustrates an embodiment of a conduit system to transport the thermal fluid to the outlet head shell **122**, according to the present disclosure. In FIG. 1D, thermal fluid can be withdrawn from the tube reactor through the port **134** in the tube reactor shell **102**. The withdrawn fluid can then pass through the conduit **148**, through the pump **150**, and into a series of additional conduits **152**. The additional conduits **152** can be in fluid communication with a number of half-pipes **154**, such that thermal fluid can enter the half-pipes **154** and provide cooling or heating to the outlet head shell **122**.

Also, as shown in FIG. 1D, thermal fluid can also be introduced to the flanges **136**, **138** so that temperature across the flange area is also equilibrated. Thermal fluid can then be withdrawn from the half-pipes **154** through the exit conduits **156** and, in the embodiment shown, circulated to a drum **158**. In some embodiments, the thermal fluid circulated to the drum **158** can be introduced to the drum **158** by means of a vertical internal standpipe to avoid hammering, as appreciated by one skilled in the art. In addition, the various conduits **154** can include various apparatuses for balancing and/or otherwise controlling the flow of thermal fluid through and/or between the conduits **154**.

FIG. 2 illustrates an embodiment of a tube reactor **200** according to an embodiment of the present disclosure. As shown in FIG. 2, the tube reactor **200** includes an exterior shell **202** that encloses a space that contains reactor tubes **204**, as discussed herein. In addition, as discussed herein, a top tubesheet and a bottom tube sheet **208** can hold the tubes **204** in position within the tube reactor shell **202**. Ports **234** enter the tube reactor shell **202** for the introduction and withdrawal of thermal fluid. Also, process gases can enter the tube reactor **200** through the inlet in an inlet head.

As shown in FIG. 2, the outlet head **216** can be removably affixed to the tube reactor shell **202** via optional flanges **236** and **238**, but it is possible that the outlet head **216** be permanently affixed to the tube reactor shell **202**. If the outlet head **216** is removable, then a method for maintaining the temperature of the outlet head **216** approximately equal to that of the tube reactor shell **202** can be used, if necessary, to prevent differential thermal expansion, as discussed herein.

As illustrated, the insert **224** can reside within the outlet head **216**, occupying a central region of the head space **212** defined by the outlet head **216**. The insert **224** can be supported in position within the outlet head **216** using any appropriate structure, such as various types of bracketing and supports.

As shown, the insert **224** can be positioned within the outlet head **216** so a space **260** exists between the outlet ends of the reaction tubes **204** and the insert **224**, to permit reaction product gases to enter the outlet head **216**. The space **260** can



separate the insert **224** from the outlet head **216**, providing a restricted path through which process gases can flow toward the outlet **220**. Reaction product gases entering the outlet head **216** can contact the top of the insert **224** and flow radially outward, in the directions of arrows **262**, where they enter the space **260** and flow downward toward the outlet **220**.

FIG. **2A** is a top-down view of the tube reactor **200** taken at line **2A** in FIG. **2**. Gas velocities through an annular space **266** can be increased relative to velocities in the absence of the insert **224**, thereby reducing residence time and reducing byproduct formation and stagnant areas. In some embodiments, the residence time can be reduced by up to forty (40) percent as compared to the outlet head **216** without the insert **224**.

In the embodiment shown, the insert **224** can be hollow or solid, however, by including a hollow insert **224**, mass and cost can be reduced as compared to a solid insert **224**. In such embodiments, the insert **224** can be filled with an inert gas, for example, nitrogen.

As shown in FIG. **2**, the outlet head **216** can include an additional insert **263** in addition to the insert **224**. In this embodiment, the additional insert **263** can be a diverter plate **264**. As used herein, a “diverter plate” refers to a non-perforated plate positioned in the outlet head space **212** to direct the flow of reaction product gases exiting the reaction tubes **204** and/or rapidly mix the reaction product gases in the outlet head space **212** to reduce the occurrence of hot spots and/or stagnant areas. In some embodiments, the outlet head **216** can include a number of diverter plates **264**.

As shown by the arrows in FIG. **2**, diverter plates **264** positioned in a substantially horizontal position below the reaction tube **204** outlets can direct the flow of reaction product gases from reaction tubes **204** on an outer periphery of the tube reactor **200** towards the insert **224**. In addition, diverter plates **265** positioned in a substantially vertical position and in an angled position below the horizontal diverter plates **264** can be perforated and can rapidly mix the reaction product gases as they flow around the horizontal diverter plates **264** and the insert **224** and through the perforated diverter plates **265**.

As discussed herein, in some embodiments, the outlet head **216** can be removably affixed to the tube reactor shell **202** in order to permit access to the interior of the tube reactor **100** for purposes such as maintenance, repair, and/or catalyst removal and/or replacement. In such embodiments, the insert **224** and/or diverter plates **264**, **265** can be permanently positioned within the outlet head **216** since the outlet head **216** can be removed when, for example, the reaction tube **204** catalyst is replaced.

In some embodiments, however, the outlet head **216** can be welded to the tube reactor shell **202**. In such embodiments, manways can be provided to allow for a person to enter the outlet head **216** to perform maintenance, repair, and/or catalyst removal and/or replacement. As one skilled in the art will appreciate, however, when the outlet head **216** is provided with enough space for a person to enter the outlet head **216**, the outlet head space **212** size increases, residence times in the outlet head **216** increase, and the risk of hot spot formation and possible consequent ignition and/or byproduct formation increases.

FIG. **3** illustrates an embodiment of a tube reactor **300** according to the present disclosure. As shown in FIG. **3**, the outlet head **316** is welded to the tube reactor shell **302**, and the outlet head **316** is large enough to fit a person **366** in the outlet head space **312**. To decrease the residence time of the reaction product gases exiting the tubes **304** in the outlet head **316**, an insert **324** can be positioned in the outlet head **316**. In such

embodiments, the insert **324** can be brought into the outlet head **316** in pieces and assembled in the outlet head **316**, as shown in FIG. **3**.

Since the insert **324** is assembled in the outlet head **316** from separate pieces, it is possible that the pieces would not join together to create a fluid-tight seal. In addition, it is possible that the pieces would not be able to sustain full-process pressure as assembled. As such, in some embodiments, an inert gas (e.g., methane, nitrogen) can be flowed into the space **312** between the insert **324** and the outlet head shell **322**, maintaining a positive pressure inside the space **312** to prevent or minimize reaction product gases from entering the space **312**.

As illustrated, the embodiment shown in FIG. **3** includes a heat exchanger **332** positioned at least partially in the outlet head **316**. The heat exchanger **332** can be used to quickly cool the reaction product gases exiting the reactor tubes **304**, as discussed herein. In some embodiments, the outlet head **316** does not include the heat exchanger **332** and the insert **324** is constructed to direct the flow of reaction product gases to the outlet **320** as quickly as possible.

FIG. **4** illustrates an embodiment of a tube reactor **400** according to embodiments of the present disclosure. As discussed herein, in some embodiments, the outlet head **416** can be permanently affixed (e.g., welded) to the tube reactor shell **402**. In such embodiments, the outlet head space **412** is large enough for a person to enter the outlet head **416** for maintenance, repairs, and/or catalyst removal and/or replacement. As such, the outlet head space **412** has an increased volume as compared to embodiments with a removable outlet head **416** with an insert positioned in the outlet head **416**.

As shown in FIG. **4**, in some embodiments, the tube reactor **400** can include an insert where the insert is a diverter plate **464**. The diverter plate **464** can be positioned directly below the bottom of the reaction tubes **404** and can have a perforated construction. As such, the reaction product gases exiting the reaction tubes **404** can be forced to flow horizontally between the bottom tubesheet **408** and the diverter plate **464**, thus achieving rapid mixing of gases between “hot-spotted” tubes **404** and normal temperature tubes **404**. The reaction product gases can also flow through a small number of perforations in the diverter plate **464** and into the outlet head space **412**. By rapidly mixing the reaction product gases, hot gas formed in a reaction tube **404** can be quickly mixed with cold gas from the other reaction tubes **404**, reducing the occurrence of hot spots.

In such embodiments, the diverter plate **464** can be formed of pieces small enough to fit through a manway **468** and can be bolted in place in the outlet head **416**. In addition, to avoid any stagnant areas in the outlet head space **412**, gas flow can be introduced into the outlet head space **412** to move the reaction product gases towards the outlet **420** in the outlet head **416**.

In some embodiments, the outlet head **416** can also include a second diverter plate **464** to redistribute the reaction product gases in the outlet head space **412**. The second diverter plate **464** can have a perforated construction, allowing the reaction product gas to flow through the diverter plate **464** and in the annular space **469** between the diverter plate **464** and the outlet head **416** thus promoting positive flow along the outlet head **416** wall and reducing the accumulation of catalyst dust.

The process gases used during operation of the foregoing embodiments of the invention can be any material or mixture of materials that is capable of reaction within a tube reactor. In some embodiments, the process gas can be a gas mixture for making an alkylene oxide, particularly ethylene oxide. An alkylene oxide-producing process gas can include at least one



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alkene (preferably ethylene), oxygen, and optionally other gases as may be desirable to provide reaction control, such as alkyl halides, NOx compounds and the like. The process can be operated at superatmospheric pressures (such as 1.01 Bar absolute to 28.5 Bar absolute). In addition, the process temperature within the tube reactor can be controlled at temperatures in a range of 50 to 310° C., although this can vary with the particular process. In the case of ethylene oxide production processes, the reaction tubes contain a suitable oxidation catalyst, for example a silver catalyst, which may be supported and may contain or be used in conjunction with one or more catalyst promoters.

It is to be understood that the above description has been made in an illustrative fashion, and not a restrictive one. Although specific embodiments have been illustrated and described herein, those of ordinary skill in the art will appreciate that other component arrangements can be substituted for the specific embodiments shown. The claims are intended to cover such adaptations or variations of various embodiments of the disclosure, except to the extent limited by the prior art.

In the foregoing Detailed Description, various features are grouped together in exemplary embodiments for the purpose of streamlining the disclosure. This method of disclosure is not to be interpreted as reflecting an intention that any claim requires more features than are expressly recited in the claim. Rather, as the following claims reflect, inventive subject matter lies in less than all features of a single disclosed embodiment. Thus, the following claims are hereby incorporated into the Detailed Description, with each claim standing on its own as a separate embodiment of the invention.

What is claimed:

1. A tube reactor comprising:

multiple, substantially parallel reaction tubes arranged within a tube reactor shell, the reaction tubes spaced apart such that a thermal fluid can flow between the reaction tubes and transfer heat between the reaction tubes and the thermal fluid during operation;

an inlet head positioned adjacent the tube reactor shell defining an inlet head space, where the inlet head space is in fluid communication with an inlet end of the reaction tubes;

an outlet head positioned adjacent the tube reactor shell including an outlet head shell that defines an outlet head space; and

at least one insert positioned within the outlet head space, where the insert slopes from an edge region towards an outlet end so that a height of the outlet head space is greater near a center of the outlet than at the edge region and the insert reduces an outlet head volume for reaction product gases by at least twenty-five percent, the outlet head volume being in fluid communication with an outlet end of the reaction tubes and in fluid communication with a reactor outlet.

2. The tube reactor of claim 1, where the outlet head volume is at least approximately 40 percent reduced in volume compared to that defined by the outlet head.

3. The tube reactor of claim 1, where the outlet head shell is at least partially thermally isolated from reaction product gases entering the outlet head space from an exit end of the reaction tubes.

4. The tube reactor of claim 1, where the outlet head is removably attached to the tube reactor shell.

5. The tube reactor of claim 1, where the tube reactor further includes a thermal fluid inlet for introducing a thermal fluid into the tube reactor shell and into contact with the

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reaction tubes and a thermal fluid outlet for withdrawing the thermal fluid from the tube reactor.

6. The tube reactor of claim 5, where the thermal fluid withdrawn from the tube reactor is contacted with the outlet head shell to maintain the outlet head shell at a temperature approximately equal to a temperature of the tube reactor shell.

7. The tube reactor of claim 1, where the outlet head includes a heat exchanger disposed at least partially within the outlet head to cool reaction product gases exiting the reaction tubes.

8. The tube reactor of claim 7, where the tube reactor includes a second heat exchanger located outside the outlet head, where the second heat exchanger is close-coupled to the heat exchanger disposed at least partially within the outlet head.

9. The tube reactor of claim 1, where the insert occupies a central portion of the space defined by the outlet head and rapidly mixes a reaction product gas exiting the reaction tubes in the outlet head space.

10. The tube reactor of claim 1, where the insert is a diverter plate.

11. The tube reactor of claim 10, where the diverter plate is formed of a number of diverter plate sections assembled into a perforated construction inside the outlet head space.

12. A method of operating a tube reactor, comprising:  
flowing process gases through multiple, substantially parallel reaction tubes arranged within a tube reactor shell, where the process gases react to form reaction product gases in the tubes;

flowing a thermal fluid into the tube reactor shell, where the thermal fluid can flow between the reaction tubes and transfer heat between the reaction tubes and the thermal fluid;

removably attaching an outlet head to the tube reactor shell, where the outlet head defines an outlet head space that is in fluid communication with an outlet end of the reaction tubes and includes an outlet head shell;

exiting the reaction product gases from the tubes into the outlet head, where the outlet head shell is at least partially thermally isolated from the reaction product gases; and

circulating the thermal fluid from the tube reactor shell through the outlet head shell to maintain the outlet head shell at a temperature approximately equal to a temperature of the tube reactor shell.

13. The method of claim 12, where the outlet head and tube reactor shell include mating flanges and removably attaching the outlet head to the tube reactor shell includes mating the flanges of the outlet head and the tube reactor shell.

14. The method of claim 12, where the method includes maintaining the mating flanges of the outlet head and the tube reactor shell at approximately the temperature of the tube reactor shell.

15. A process, comprising:  
flowing process gases through a tube reactor under conditions such that a chemical reaction occurs within reaction tubes in the tube reactor to form a desired reaction product gas, wherein said tube reactor includes:

multiple, substantially parallel reaction tubes arranged within a tube reactor shell, where the reaction tubes are spaced apart such that a thermal fluid can flow between the reaction tubes and transfer heat between the reaction tubes and the thermal fluid during operation, and

an outlet head removably attached to the tube reactor shell and which defines an outlet head space in fluid communication with an outlet end of the reaction

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tubes and a reactor outlet, the outlet head including an outlet head shell at least partially thermally isolated from process gases contained in the outlet head space; where the temperature of the outlet head shell is maintained approximately equal to that of the tube reactor shell.

**16.** The process of claim **15**, where the tube reactor includes at least one insert positioned within the outlet head space, where the insert slopes from an edge region towards an outlet end so that a height of the outlet head space is greater near a center of the outlet than at the edge region and the insert reduces an outlet head volume for reaction product gases by at least twenty-five percent, the outlet head volume being in

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fluid communication with an outlet end of the reaction tubes and in fluid communication with a reactor outlet.

**17.** The process of claim **15**, where the outlet head volume is at least approximately 40 percent reduced in volume compared to that defined by the outlet head.

**18.** The tube reactor of claim **6**, where the outlet head shell includes a jacket and/or a conduit through which the thermal fluid from the tube reactor shell can flow to maintain the temperature of the outlet head shell approximately equal to a temperature of the tube reactor shell.

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